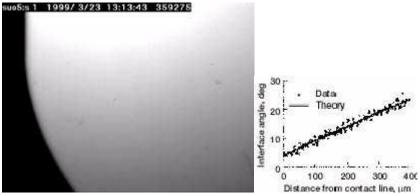
## How Does a Liquid Wet a Solid? Hydrodynamics of Dynamic Contact Angles

A contact line is defined at the intersection of a solid surface with the interface between two immiscible fluids. When one fluid displaces another immiscible fluid along a solid surface, the process is called dynamic wetting and a "moving" contact line (one whose position relative to the solid changes in time) often appears. The physics of dynamic wetting controls such natural and industrial processes as spraying of paints and insecticides, dishwashing, film formation and rupture in the eye and in the alveoli, application of coatings, printing, drying and imbibition of fibrous materials, oil recovery from porous rocks, and microfluidics.

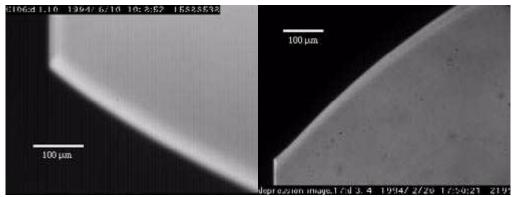


Left: Shadowgraph of the static meniscus of silicone oil on Pyrex (Corning, Corning, NY). Field of view is about 600 µm in the horizontal direction. The fluid slightly overfills a Teflon beaker, 10 cm in diameter. A Pyrex tube, 2.54 cm in diameter, is immersed in the fluid. Because of the slight overfill, the meniscus appears above the beaker rim and can, thus, be imaged optically. Right: Static meniscus of silicone oil on Pyrex: The angle between the solid and the interface tangent is shown versus the distance from the contact line from image analysis of the picture in the first figure. The slope extrapolates to a static contact angle, ~4°, at the contact line. The solid line shows the best fit of the static capillary theory. The theory's only adjustable parameter is the contact angle.

The contact angle, the angle formed at the intersection of the solid and the fluid-fluid interfaces, is a key material property needed to determine the shape of the fluid-fluid interface. It serves as the boundary condition for a differential equation describing the interface shape. In static capillary systems (e.g., the water meniscus formed on a clean glass plate), the static contact angle is routinely used for this purpose. The preceding image shows a magnified view near the contact line of a static meniscus formed by polydimethylsiloxane (silicone oil) on clean Pyrex: the air is clear, the fluid below is dark, and the vertical straight line at the top right is a Pyrex glass immersed in the fluid. Since silicone oil wets glass "perfectly," its contact angle is very close to zero. The preceding graph shows the angle between the solid and the tangent to the static interface of the

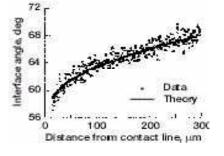
shadowgraph image, as a function of the distance to the contact line, r. The solid line is the solution of the static capillary theory (ref. 1). Clearly, the slope may be safely extrapolated to the contact line, r = 0, at which point the angle equals the static contact angle of the system.

In dynamic systems, however, the "dynamic" contact angle is not well defined. For example, when a silicone oil of viscosity  $\mu = 10$  poise (1000 times more viscous than water) and surface tension  $\sigma = 20$  dyn/cm advances on Pyrex with velocity U = 0.02 cm/sec, the following image on the left shows that the dynamic contact angle is close to 67°. The controlling factor for the observed dynamic contact angle is the dimensionless group known as the capillary number,  $Ca = U\mu/\sigma$ . When Ca = 0.1, silicone oil forms a contact angle close to  $120^{\circ}$  (right image).

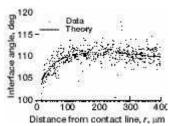


Dynamic interface shape of silicone oil on Pyrex. Left: Ca, 0.01. Right: Ca, 0.1. Because of the high viscous forces near the contact line at Ca = 0.1, the macroscopic meniscus is bent into depression. Thus, the liquid is clear and the air is dark.

Despite their well-behaved appearance, these dynamic interfaces are fundamentally different from static interfaces. The final two graphs show that, when the dynamic interface shape is digitized and the angle is plotted versus the distance from the contact line, the slope does not attain a well-defined limit at the contact line (r = 0). Viscous forces dramatically bend the interface near the contact line. This suggests that, in contrast to the static contact angle (see the first graph, preceding page), the dynamic contact angle is not a well-defined quantityit is not at all clear where on the interface one should apply the slope condition in order to calculate the interface shape. Nevertheless, a dynamic contact angle is still necessary to calculate the interface shape in dynamic conditions, the pressure drop necessary to move a meniscus in a capillary tube, and the spreading dynamics of small droplets.



Angle between solid and tangent to the interface versus distance from contact line at Ca = 0. The solid line shows the best fit of the dynamic theory (ref. 2). The theory's only adjustable parameter is a property that controls dynamic wetting and whose role is that of an "apparent" contact angle.



Angle between solid and tangent to the interface versus distance from contact line at Ca = 0. The solid line shows the best fit of the dynamic theory (ref. 2).

Principal investigator Professor Stephen Garoff of Carnegie Mellon University and coinvestigator Enrique Ramé of the National Center for Microgravity Research in Fluids and Combustion have been studying these systems for about 10 years. The objectives are

- 1. Identify and measure material-dependent, geometry-independent properties for describing dynamic wetting predicted in theoretical analyses valid at a low *Ca*.
- 2. Test the theories by measuring the property in two different geometries and using it to predict the flow in a third geometry.
- 3. Generate geometry-independent dynamic contact angle information outside the range of the validity of the theory  $(Ca \sim 1)$ .

By using an asymptotic theory valid at Ca << 1 (ref. 2) and measurements of the interface near the contact line, we can extract a material parameter describing wetting dynamics. This parameter can be translated to geometries different from that where the measurement was performed, giving the approach predictive power. When Ca is not << 1, a geometry-free region of flow near the contact line must first be identified; then the interface shape and velocity field measured in that region can be used as boundary conditions for actual calculations. During the last year, as part of our ground-based program, we have begun to understand some subtleties of unsteady wetting behavior through a series of controlled experiments; a manuscript describing this work is in preparation.

The NASA Glenn Research Center is in the process of developing flight hardware to conduct a microgravity experiment to study the microscale phenomena in the vicinity of the moving contact line. In the absence of gravity, the region dominated by capillary force is enlarged, allowing detailed observations of flow and meniscus shape.

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